

## Synthesis, Crystallographic Characterization and Computational Study of Two Polymorphs of 5,6-Dimethyl-2-(pyridin-2-yl)-1-[(pyridin-2-yl)methyl]-1*H*benzimidazole

The title compound was prepared by the condensation of 1,2-diamino-4,5-dimethylbenzene and 2-pyridinecarbaldehyde in a forty percent yield. Single crystals of polymorph I were obtained by vapor diffusion of hexane into an ethanolic solution of the benzimidazole, whereas single crystals of polymorph II were obtained by vapor diffusion of diethyl ether into a DMF solution of the benzimidazole. I crystallizes in the monoclinic space group C2/c and II crystallizes in the orthorhombic space group Pbca, each with one molecule in the asymmetric unit. Although the bond distances and angles displayed by the two molecules are virtually identical, the stereochemistry differs. In I, the 2-(pyridine-2-yl) substituent is canted by only 2.75 (11)°; however, in II, the angle is 30.68 (4)°. Both I and II are stabilized by weak intramolecular hydrogen-bonding interactions between the 1-[(pyridine-2-yl)methyl] and 2-(pyridine-2-yl) substituents. DFT calculations were employed to determine the basis set superposition corrected-energy (counterpoise method) of intermolecular hydrogen bonding interaction energies between polymorph pairs. In addition, a conformational analysis of the 1-[(pyridine-2-yl)methyl] and 2-(pyridine-2-yl) substituents were performed for both I and II.

*Keywords:* Benzimidazole; pharmaceutical compound; crystal structure; polymorphs; density functional theory; DFT; computational chemistry.